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International filing date (day/month/year) 15 January 2000 (15.01.00)	**Priority date** (day/month/year) 27 January 1999 (27.01.99)
Applicant YAMAMOTO, Tetsu	

1. The designated Office is hereby notified of its election made:

[X] in the demand filed with the International Preliminary Examining Authority on:

17 August 2000 (17.08.00)

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IMPORTANT NOTIFICATION

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1. The following indications appeared on record concerning:

[X] the applicant [ ] the inventor [ ] the agent [ ] the common representative

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[ ] the person [X] the name [ ] the address [ ] the nationality [ ] the residence

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METHOD FOR PRODUCING BRIDGED POLYMER MEMBRANE AND FUEL CELL

Tetsu Yamamoto

INTERNATIONAL APPLICATION

IN ENGLISH

-with- Two (2) Sheets of Drawings

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(57) Abstract

A method for producing a bridged polymer membrane includes the steps of: obtaining a liquid medium comprising a basic polymer having an amino group in a repeating unit, a bridging agent, and a solvent; shaping the liquid medium into a membrane configuration to obtain the shaped membrane; and bridging the basic polymer by the bridging agent in the shaped membrane. A fuel cell has the bridged polymer membrane. The mechanical strength of the polymer electrolyte membrane is improved.

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Method for producing bridged polymer membrane and fuel cell

Field of the Invention

The present invention relates to a method for producing a bridged polymer membrane and a fuel cell.

Related Art

A cell includes an electrolyte and a pair of electrodes separated by the electrolyte. In case of a fuel cell, a fuel such as a hydrogen gas is supplied to one of the electrodes, and an oxidizing agent such as an oxygen gas is supplied to the other electrode, thereby converting chemical energy involved in the oxidation of the fuel into electric energy. The electrolyte permeates hydrogen ions, that is, protons but does not permeate reactive gases such as the hydrogen gas and the oxygen gas. Typically, a fuel cell has a plurality of single cells, and each of the single cells has an electrolyte and a pair of electrodes separated by the electrolyte.

As the electrolyte for the fuel cell, a solid such as a polymer electrolyte membrane and a liquid such as phosphoric acid are used. Recently, the polymer electrolyte membranes have been receiving attention as the electrolytes for the fuel cell. For example, perfluorosulfonic acid polymers and complexes between basic polymers and strong acids are used as materials for the polymer electrolyte membranes.

Typically, the perfluorosulfonic acid polymer has a framework of perfluorocarbon such as a copolymer of tetrafluoroethylene and trifluorovinyl and a side chain being bonded thereto and having a sulfonic acid group such as a side chain that a sulfonic acid group is bonded to perfluoroalkylene group. The sulfonic acid group releases a hydrogen ion to convert into an anion, and therefore conducts proton.

Polymer electrolyte membranes comprising complexes of basic polymers and strong acids have been developed. International Publication WO96/13872 and its corresponding U.S. Pat. No. 5,525,436 disclose a method for producing a proton conductive polymer electrolyte membrane by immersing a basic polymer such as polybenzimidazoles in a strong acid such as phosphoric acid, sulfuric acid and so on. The fuel cell employing such a polymer electrolyte membrane has the advantage that it can be operated at 100 °C or above.

J. Electrochem. Soc., Vol. 142, No. 7, 1995, ppL121-L123 describes that immersing

a polybenzimidazole in 11M phosphoric acid for at least 16 h impregnates five molecules of phosphoric acid per unit of the polybenzimidazole.
Further, International Publication WO97/37396 and its corresponding U.S. Pat. No. 5,716,727 describe a method for producing a polymer electrolyte membrane by obtaining a solution of a polybenzimidazole dissolved in trifluoroacetic acid, followed by adding phosphoric acid to the solution, and subsequently by removing the solvent.

The whole disclosures of WO 96/13872, *J. Electrochem. Soc.*, Vol. 142, No. 7, 1995, ppL121-L123 and WO97/37396 are incorporated herein as reference.
Even though a basic polymer by itself has a sufficient mechanical strength, there are cases that the mechanical strength of the basic polymer decreases to an insufficient degree by impregnating the basic polymer with a strong acid for providing proton conductivity. Therefore, it is desired to further improve the mechanical strength of the basic polymer for applying the complex of the basic polymer and the strong acid to the electrolyte membrane for the fuel cell and so on.
U.S. Pat. No. 5,599,639 describes a basic polymer wherein a sulfonic acid group is introduced into a basic polymer such as polybenzimidazole and so on through a linker such as an alkylene and so on. The basic polymer is required to incorporate water therein for providing proton conductivity so that the basic polymer may be used as the electrolyte membrane for the fuel cell.

However, when the basic polymer is immersed in water, there are cases that the basic polymer swells and that a sufficient mechanical strength is not achieved. The whole disclosure of U.S. Pat. No. 5,599,639 is incorporated herein as reference.
The present inventors tried to improve the mechanical strength by shaping a basic polymer into a membrane configuration followed by reacting with an external bridging agent. However, the basic polymer in a gel or solid form did not smoothly react with the external bridging agent.

Summary of the Invention

To solve the aforementioned problem, the present invention is accomplished by shaping a basic polymer into a membrane configuration followed by proceeding a bridging reaction.

According to one aspect of the present invention, there is provided a method for producing a bridged polymer membrane, comprising the steps of: obtaining a liquid

medium comprising a basic polymer having an amino group in a repeating unit, a bridging agent, and a solvent; shaping the liquid medium into a membrane configuration to obtain a shaped membrane; and bridging the basic polymer by the bridging agent in the shaped membrane.

Preferably, the bridging agent has at least two epoxy groups or isocyanate groups in the molecule thereof.

Preferably, the liquid medium contains 0.001 to 0.8 mole of the bridging agent per unit of the basic polymer.

Preferably, the basic polymer is selected from the group consisting of polybenzimidazoles, polyimidazoles, polyvinylimidazoles, and polybenzbisimidazoles.

Preferably, the method may further comprise the step of impregnating the basic polymer with a strong acid for providing proton conductivity. The strong acid may be impregnated in the form of the liquid medium. Alternatively, the strong acid may be impregnated after the shaping but before the heating. Alternatively, the strong acid may be impregnated after heating.

Alternatively, the basic polymer may have a strong acid group in the repeating unit in the basic polymer. The presence of the strong acid provides proton conductivity.

According to another aspect of the present invention, there is provided a fuel cell comprising a plurality of single cells, each of the single cells comprising a bridged polymer membrane obtained by the aforementioned method and a pair of electrodes sandwiching the bridged polymer membrane.

Brief Description of the Drawings

Fig. 1 is a cross section of a part of a fuel cell.

Fig. 2 is an enlarged portion A of Fig. 1.

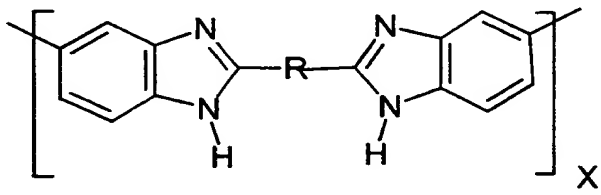
Preferred Embodiments of the Invention

The present invention may include the step of obtaining a liquid medium comprising a prescribed basic polymer, a bridging agent, and a solvent. Typically, the basic polymer is dissolved in the solvent to obtain a solution, and the bridging agent is added to the solution, followed by mixing thereof.

As the basic polymer, the basic polymer including an amino group in a repeating unit is used. Since the amino group is present in the repeating unit, the polymer becomes basic and the amino group may react with the bridging agent. In view of the reactivity with the bridging agent, the amino group in the repeating unit is preferably a primary amino group or a secondary amino group.
The repeating unit in the basic polymer preferably contains an aromatic ring containing at least one nitrogen atom. The aromatic ring is preferably a five-membered ring or a six-membered ring containing one to three nitrogen atoms, which may be fused with another ring, particularly another aromatic ring.

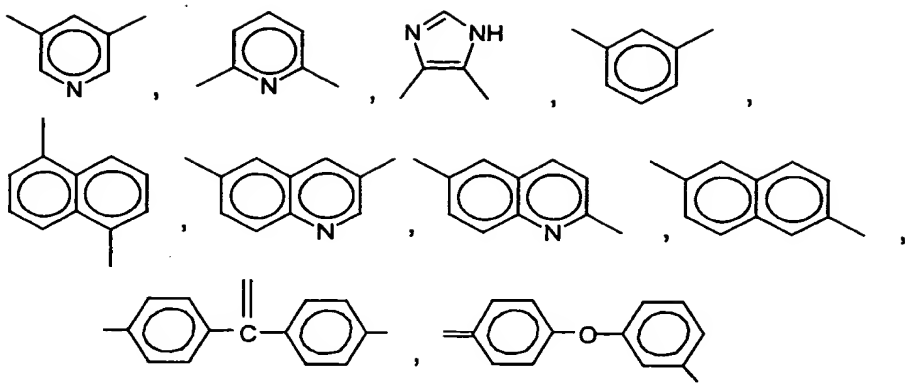
Preferably, the basic polymer is soluble to the solvent. Specifically, a solution preferably dissolves not less than 1 percent by weight of the basic polymer and further preferably not less than 2 percent by weight. The features facilitates forming a uniform polymer membrane without forming pores therein.
The basic polymer which may be used in accordance with the present invention includes polybenzimidazoles, polyimidazoles, polyvinylimidazoles, and polybenzbisimidazoles. Among these, polybenzimidazoles are preferable.

As for the polybenzimidazoles, those of the following formula are preferably used:



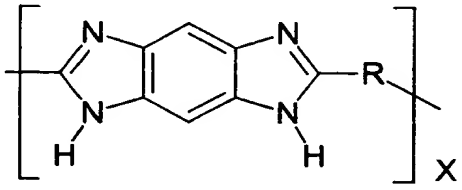
(I)

wherein R represents alkylene, perfluoroalkylene, or a substituent of any of the following formulae:



further wherein each of alkylene and perfuoroalkylene groups, which may be R, has 1 to 10 carbons preferably, and more preferably 1 to 6 carbons.

Furthermore, the basic polymers which may also be used are polybenzbisimidazoles of the following formula:



wherein R has the aforementioned meaning.

A strong acid group such as a sulfonic acid group(-SO_3H), a phosphate monoester group(-O-P(=O)$(OH)_2$) and so on may be introduced into the amino group of the polybenzimidazoles or the polybenzbisimidazoles through a linker.

As for the linker, the aforementioned R may be used. Alternatively, the linker may include a linear or branched hydrocarbon group having 1 to 20 carbon atoms, which may be substituted with a fluorine atom, which may be interrupted by an oxygen atom (-O-) or a group represented by a formula –N(R^2)- wherein R^2 is a hydrogen atom or a lower alkyl group having 1 to 6 carbon atoms. The hydrocarbon group includes: a lower alkylene group having 1 to 20 carbon atoms, which may be interrupted by an oxygen atom or an arylene group such as a phenylene group, and which may be branched; and an arylene group such as a phenylene group, which may be substituted with a lower alkyl group, a lower alkoxy group, a sulfonic acid group, an amino group, a fluorine atom and so on. Alternatively, a group represented

by a formula $-(CR^3R^4)_p-O-(CR^5R^6)_q-$

wherein p and q are independent, and each is an integer of 1 to 10; and

R^3, R^4, R^5, and R^6 are independent, and each is the same or different and a hydrogen atom; a fluorine atom; a lower alkyl group or a lower alkoxy group having 1 to 6 carbon atoms; an aryl group such as a phenyl group which may be substituted with a lower alkyl group, a lower alkoxy group, a sulfonic acid group, an amino group, a fluorine atom and so on; or a strong acid group such as a sulfonic acid group, a phosphate monoester group, and preferably a hydrogen atom; a fluorine atom; or a lower alkyl group having 1 to 6 carbon atoms.

For example, a group represented by a formula $>N-(CR^3R^4)_r-SO_3H$ wherein N represents a nitrogen atom in the imidazole ring of the polybenzimidazoles or the polybenzbisimidazoles; r is an integer of 1 to 20; and R^3 and R^4 have the aforementioned meanings may be introduced into the polybenzimidazoles or the polybenzbisimidazoles.

When a strong acid group is introduced into the nitrogen atom of the polybenzimidazoles or the polybenzbisimidazoles through the linker, it is not necessary that the linker and the strong acid are introduced into all of the nitrogen atoms. The linker and the strong acid may be introduced into some of the nitrogen atoms, and hydrogen atoms may remain bonded to the other nitrogen atoms. The remaining nitrogen atoms may be reacted with the bridging agent, and therefore it is preferable.

For example, the linker and the strong acid may be introduced into 5 to 85 percent of the nitrogen atoms of the basic polymer such as the nitrogen atoms of the imidazole ring, and particularly, the linker and the strong acid may be introduced into 10 to 75 percent of the nitrogen atoms, and further particularly, the linker and the strong acid may be introduced into 15 to 40 percent of the nitrogen atoms.

The basic polymer may be reacted with a sultone in a solution of the basic polymer to sulfonate or sulfoalkylate the basic polymer. In the reaction, for example, a solution of 1 to 30 percent by weight of the basic polymer may be used, and particularly, a solution of 5 to 20 percent by weight of the basic polymer may be used. As for the solvent for the sulfonation or sulfoalkylation, the solvent for the liquid

medium, which will be described later, is preferably used.

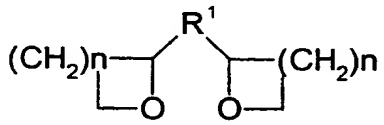
The reaction is, for example, described in U.S. Pat. No. 5,599,639, U.S. Pat. No. 4,814,399 and Ahmed Mstafa, Chemical Review, pp. 195-223(1954), and these documents are incorporated herein as reference.

As the solvent used for the liquid medium, the solvent that does not react with the bridging agent is preferable. An organic solvent is preferable, and an organic solvent having polarity is further preferable. The organic solvent includes, for example, lower amides such as N-methyl-2-pyrrolidone, N, N-dimethylacetamide, dimethylformamide; dimethylsulfoxide and so on. A mixture of these solvents may be used.

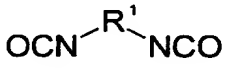
The liquid medium may be either a solution or a suspension. In case of the solution, the basic polymer is dissolved in the solvent. In case of the suspension, particles of the basic polymer serving as a dispersion phase is dispersed in a solvent serving as a continuous phase. Alternatively, the liquid medium may be a slurry or a paste.

The bridging agent is not limited as long as the bridging agent includes a compound having a functional group reacting with an amino group. The bridging agent preferably has at least two functional groups for reacting with the amino group in the molecule thereof, and the bridging agent is typically an organic compound. Examples of such functional groups include epoxy groups and isocyanate groups. However, when the epoxy group and the isocyanate group are present in the molecule of the bridging agent, both groups react each other and therefore, it is not preferable.

The organic compound having not less than two epoxy groups and not less than two isocyanate groups includes, for example, an epoxy compound represented by formula (II), and an organic compound represented by formula (III). Further, the hydrogen atoms in the epoxy groups of the epoxy compound represented by formula (II) may be substituted by a halogen or a lower alkyl group.



(II)

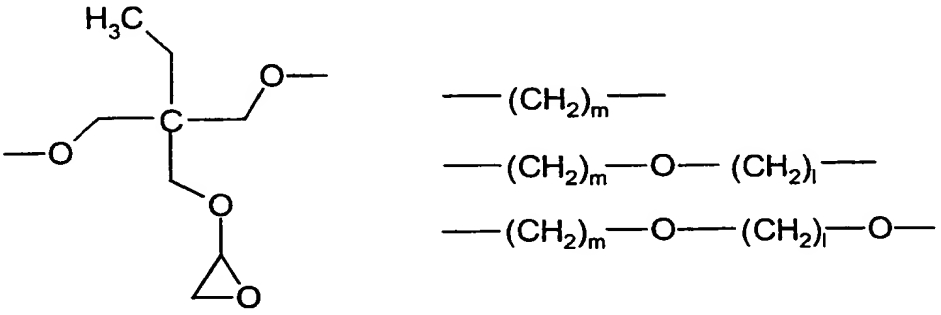


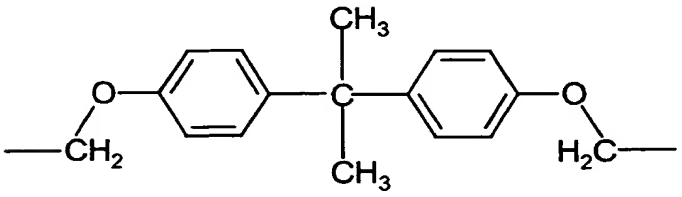
(III)

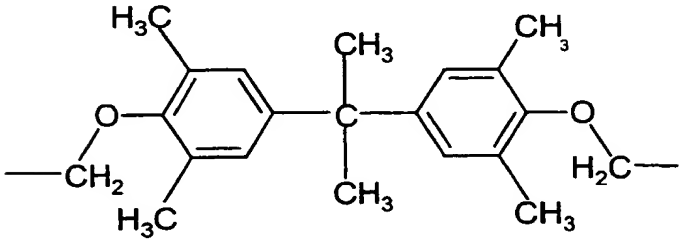
In the aforementioned formula, R^1 is a hydrocarbon group, including, for example, a linear chain or branched lower alkylene group which may be substituted by a nitro

group, an oxygen atom, an epoxy group or an aryl group; and a linear chain or branched lower alkoxy group which may be substituted with a nitro group, an epoxy group or an aryl group. As for the aryl group, a phenyl group, a naphthalenyl group and an indenyl group can be mentioned. n is an integer of 0 or 1, and preferably 0.

Examples of R^1 include the following groups.

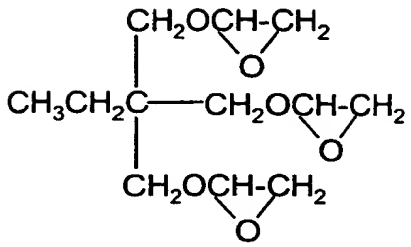






In the aforementioned formulae, m and ℓ are the same or different and each is an integer of 1 to 6.

An example of the bridging agent having three functional group in the molecule, which reacts with an amino group is shown as follows.



Preferably, the liquid medium contains at least 1 percent by weight of the basic polymer, and further preferably, the liquid medium contains at least 2 percent by weight of the basic polymer. Preferably, the liquid medium contains not more than 95 percent by weight of the basic polymer, and further preferably, the liquid medium contains not more than 80 percent by weight of the basic polymer, and particularly preferably, the liquid medium contains not more than 50 percent by weight of the basic polymer.

Preferably, the liquid medium contains 0.001 to 0.8 mole of the bridging agent per unit of the basic polymer, and further preferably, the liquid medium contains 0.01 to 0.5 mole of the bridging agent, and particularly preferably, the liquid medium contains 0.05 to 0.3 mole of the bridging agent. When an amount of the bridging agent is too much, it becomes difficult to impregnate a strong acid in the basic polymer. On the other hand, when an amount of the bridging agent is too small, the mechanical strength of the polymer membrane is not sufficiently improved.

Subsequently, the liquid medium is shaped into a membrane configuration to obtain a shaped membrane. For example, the liquid medium is cast onto a planar substrate, and then dried in a reduced pressure. Alternatively, the basic polymer may be shaped into the membrane configuration by a doctor-blade method.

The substrate for the casting or the doctor-blade method may be a semiconductor such as a silicon wafer, a synthetic polymer such as poly(ethyleneterephthalate), and a metal such as a stainless steel. When a roll made of, for example, poly(ethyleneterephthalate) is used as the substrate, a continuous castor may be used to produce a shaped membrane having a constant width and any thickness. In this case, the liquid medium is cast onto the substrate, and go through a gap having a prescribed distance, and then go through a drying furnace for drying by a warm wind.

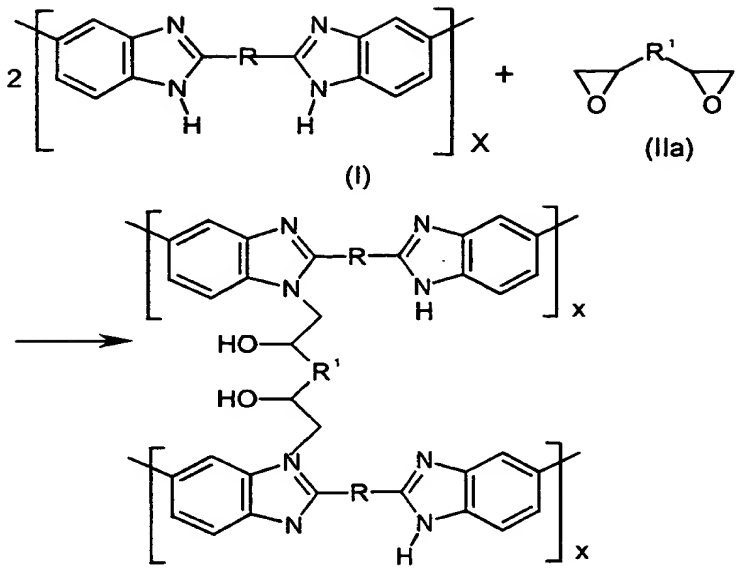
Alternatively, the basic polymer may be shaped into the membrane configuration according to the method as described in Japanese Patent Appln. Hei 10-125560, filed May 8, 1998: entitled "Method for Producing Polymer Electrolyte Membrane for Fuel Cell and Fuel Cell." Specifically, the liquid medium is poured into a cylinder having the inner surface having a cylindrical configuration, and subsequently, the cylinder is rotated. At that point, the solvent is allowed to evaporate through centrifugal force by the rotation; concurrently, a polymer membrane having a cylindrical configuration of substantially uniform thickness is formed on the inner surface of the cylinder. Thereafter, the polymer membrane having a cylindrical configuration is cut out to yield a shaped membrane having a membrane configuration. This method allows to form the basic polymer having a uniform matrix. The disclosure of Japanese Patent Appln. Hei 10-125560 is incorporated herein as reference.

In the present invention, the basic polymer is bridged by the bridging agent in the shaped membrane. Typically, the shaped membrane is heated to bridge the basic polymer by the bridging agent. However, it is not limited to heating, and a photochemical reaction may bridge the basic polymer by the bridging agent.

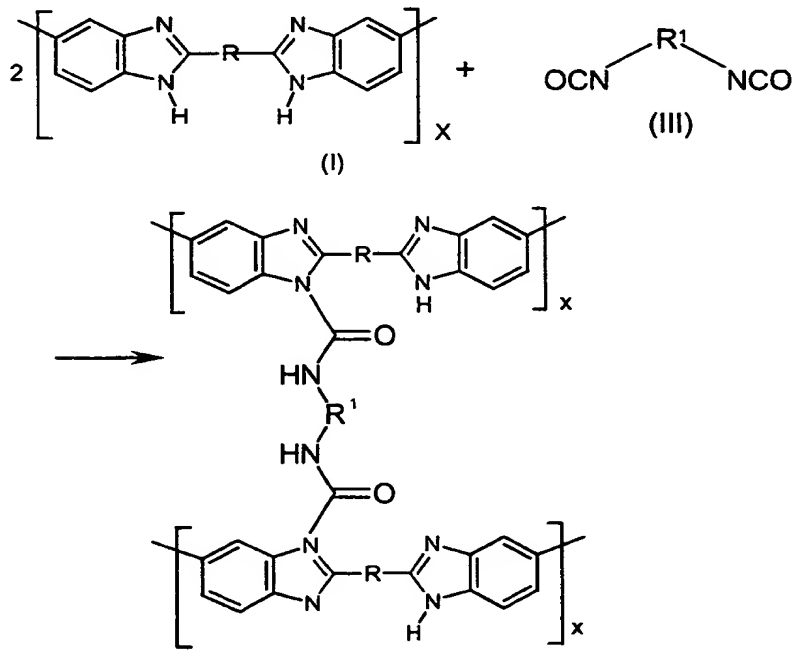
In the following description, an embodiment that the bridging agent is reacted by heating is mainly described. The aforementioned shaping step, in many cases, may include the step of heating the membrane for drying the shaped membrane. The heating for drying may form the shaped membrane and simultaneously bridge the basic polymer by the bridging agent in the basic polymer. To facilitate drying, it may be heated in a reduced pressure of not more than 1 atmospheric pressure, preferably not more than 0.1 atmospheric pressure and further preferably not more than 0.05 atmospheric pressure.

Alternatively, the heating for drying the shaped membrane may be determined to a temperature which is lower than the reaction temperature of the bridging agent, and subsequently, the shaped membrane may be distinctly heated for bridging the bridging agent. A warm wind may be used for the heating for drying or the heating for bridging.

For example, the epoxy compound represented by formula (IIa) is reacted with polybenzimidazole represented by formula (I) to bridge a polymer chain.



In the aforementioned formulae, R^1 is the same as defined above.
Similarly, the isocyanate compound represented by formula (III) is reacted with polybenzimidazole represented by formula (I) to bridge a polymer chain.



In the aforementioned formulae, R^1 is the same as defined above.

In the aforementioned formulae, it is described such that different polymer chains are

bridged for convenience of the description. However, the same polymer chain may be bridged, and an inside of one repeated unit may be bridged.

In the present invention, preferably, the step of impregnating the basic polymer with a strong acid for providing proton conductivity may be included. The strong acid may be impregnated in the form of the liquid medium. Alternatively, the strong acid may be impregnated after the shaping but before the heating. Alternatively, the strong acid may be impregnated after heating.

The embodiment that strong acid is impregnated in the form of the liquid medium refers to adding a strong acid to the liquid medium. Preferably, the bridging agent does not react with the strong acid.

Preferably, the strong acid may be impregnated after heating the shaped membrane. The bridged, basic polymer has improved mechanical strength, and the handling thereof is easier. Since the bridging agent is already reacted, and therefore, the reaction of the strong acid with unreacted bridging agent is very limited.
The bridged, basic polymer membrane may be immersed in the strong acid so that the strong acid is impregnated in the basic polymer membrane. The basic polymer may be immersed in a strong acid having a high concentration at a temperature of not less than 35°C, preferably not less than 40°C, further preferably not less than 50° for a period of not more than 5 hours, preferably not more than 1 hour. By carrying out the immersion step at 35°C or above, it becomes possible to shorten the time for immersing the basic polymer in the strong acid. However, in view of the stability of the basic polymers and the safety precautions required to handle the strong acids at high temperatures, preferably, the immersion step is carried out at not more than 200°C or below, more preferably at 100°C or below, and most preferably at 80°C or below.

Protic strong acids may be used as the strong acid. For example, phosphoric acid and sulfuric acid are preferably used.
As used in the present specification, the "phosphoric acid" includes phosphorous acid (H_3PO_3), orthophosphoric acid (H_3PO_4), pyrophosphoric acid ($H_4P_2O_7$), triphosphoric acid ($H_5P_3O_{10}$), and metaphosphoric acid. The phosphoric acid, particularly orthophosphoric acid, preferably has a concentration of not less than 80 percent by weight; more preferably, a concentration of not less than 90 percent by weight; even more preferably, a concentration of not less than 95 percent by weight;

and most preferably, a concentration of not less than 98 percent by weight. This is because the basic polymer can be impregnated with a larger number of strong acid molecules as the concentration of the strong acid increases.

The polymer electrolyte membrane obtained by the present invention, namely, the complex between the basic polymer and the strong acid, is proton conductive, and therefore, it may preferably be used as an electrolyte for cells. Nevertheless, the polymer electrolyte is not be limited to its use for the cell; but it can also be used as the electrolyte for a display element, an electrochromic element or a various sensor.

According to another aspect of the present invention, the polymer electrolyte membrane can preferably be used in the single cell for a fuel cell.

In referring to Fig. 1, the single cell for a fuel cell 10 has a polymer electrolyte membrane 12 and a pair of electrodes 20 sandwiching the polymer electrolyte membrane 12. Each of the electrode 20 has a catalytic layer 14 and a gas diffusion layer 22 for supplying a reaction gas with the catalytic layer 14.
In Fig. 2, the catalytic layer 14 has a matrix 15 consisting essentially of a polymer electrolyte membrane and a plurality of catalytic particles 16 being dispersed in the matrix. The matrix 15 along with the polymer electrolyte membrane 12 forms a channel for conducting hydrogen ions. The material for the matrix 15 is preferably the same as the material for the polymer electrolyte membrane 12. However, these materials may be different. The matrix 15 may be porous so as to allow the reactive gas to pass through. The catalytic particles preferably contact each other, thereby forming a channel for conducting electrons.

Each of the catalytic particles 16 has an electrically conductive carrier 17 and a catalytic substance 18 being loaded onto a surface of the electrically conductive carrier 17. The electrically conductive carrier may be carbon particles. The catalytic substance 18 may be platinum metal or a platinum alloy. In Fig. 2, the catalytic substance 18 is coated onto the surface of the electrically conductive carrier 17. Alternatively, the catalytic substance 18 may have a particle configuration.

The gas diffusion layer 22 is porous so as to allow the reactive gas to pass through. In Fig. 2, the gas diffusion layer 22 is made of a plurality of electrically conductive particles 26 forming a gap 24. The electrically conductive particles may be carbon particles and same as electrically conductive carrier 17. Alternatively, the electrically

conductive particle 26 is replaced by electrically conductive substance such as carbon fibers.

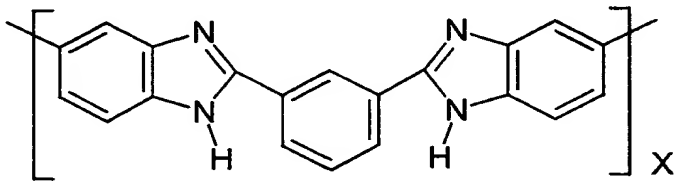
The polymer electrolyte membrane in accordance with the present invention may be used as the electrolyte membrane 12. Moreover, the electrolyte membrane 12 as well as a precursor for a single cell having one of or both catalytic layers 14 may be produced. Furthermore, the single cell may be produced by fixing the gas diffusion layer 22 to the precursor.

Examples

The following examples are merely illustrative of the present invention, and are not to be construed as limitations thereof.

Example 1

Polybenzimidazole having the structural formula described below and having an intrinsic viscosity of 1.1 (available from Hoechst Celanese Inc.) was dissolved in N,N-dimethylacetamide to yield a solution having a resin concentration of 10 percent by weight.



Subsequently, an amount shown in Table 1 of ethyleneglycoldiglycidylether as a bridging agent is added to the solution and mixed thereof. The mixed solution thus obtained was cast onto a 6-inch silicon wafer, and dried at 90 °C for four hours in a reduced pressure (10 mmHg) for forming a membrane and for proceeding the bridging reaction at the same time to obtain a cast film having a thickness of about 50 micrometer. The film thus obtained is flexible compared to unbridged polybenzimidazoles. The solubility of the cast film at room temperature in N,N-dimethylformamide was determined. The results are shown in Table 1.

Table 1

Sample	Added Amount of Ethyleneglycoldiglycidylether (molar percent)	solubility in N,N-dimethylacetamide
1	100	insoluble, no swelling
2	50	insoluble, no swelling
3	15	insoluble, swelling
4	10	insoluble, swelling
5	5	insoluble, swelling considerably
6	0	soluble

Example 2

The bridged polybenzimidazole obtained in Example 1 (50 micrometer thickness) was cut out in 3-cm square pieces. The films were washed with water to wash away the remaining N,N-dimethylacetamide. Then, they were dried at a reduced pressure and the weights of the films were measured.

Thereafter, the dried polybenzimidazole films were placed in sample vials. To each of these was added 30 ml of 85 percent by weight aqueous orthophosphoric acid, and then immersed at room temperature for 72 hours. Subsequently, the polybenzimidazole films impregnated with orthophosphoric acid were removed from the phosphoric acid, and excess phosphoric acid on their surfaces was thoroughly wiped off with filter papers. Subsequently, the weight increments were determined by weighing. After weighing, the polybenzimidazole films were placed in 1-liter volumetric flasks, and deionized water was filled up to the measuring lines and stirred. Orthophosphoric acid was extracted from the polybenzimidazole films to obtain aqueous phosphoric acid solutions. The aqueous phosphoric acid solutions thus obtained were titrated with 0.02 N sodium hydroxide solution, and the quantities of orthophosphoric acid being impregnated in the polybenzimidazole films were determined. The difference between the weight increment after impregnation with orthophosphoric acid and the weight of orthophosphoric acid of impregnation was calculated to be the quantity of the water that had been adsorbed to each polybenzimidazole film impregnated with orthophosphoric acid. These results are shown in Table 2.

Table 2

Sample	Added Amount of Ethyleneglycol-diglycidylether (molar %)	Number of phosphoric acid molecules impregnated per repeating unit of the polymer	Number of water molecules impregnated per repeating unit of the polymer
1	100	0	—
2	50	0	—
3	15	6.59	9.83
4	10	6.61	11.83
5	5	7.74	13.08

The results show that the bridged polymer membranes that is impregnated with orthophosphoric acid are obtained. Therefore, such polymer membrane may be sued as an electrolyte membrane for a fuel cell.

Even though an added amount of the bridging agent is at least 50 percent by mole, it is possible to impregnate phosphoric acid by increasing temperatures of the phosphoric acid to 50 °C. However, in this case, it takes at least seven days for impregnation, and the mechanical strength after impregnation decreases, and the film became brittle.

Example 3

Polybenzimidazole having the structural formula described in Example 1 and having an intrinsic viscosity of 1.1 (available from Hoechst Celanese Inc.) was dissolved in N,N-dimethylacetamide to yield a solution having a resin concentration of 10.0 percent by weight. Subsequently, 10 percent by mole based on a unit of polybenzimidazole of ethyleneglycoldiglycidylether as a bridging agent is added to the solution and mixed thereof.

65 gram of the solution was introduced into a stainless- steel cylinder having a cylindrical configuration with an inner diameter of 141 millimeter and a length of 408 millimeter. The cylinder was rotated at 90°C for two hours at 2500 rpm to obtain a bridged membrane of polybenzimidazole having a cylindrical configuration. A thickness of the polybenzimidazole membrane was determined at any six points. An

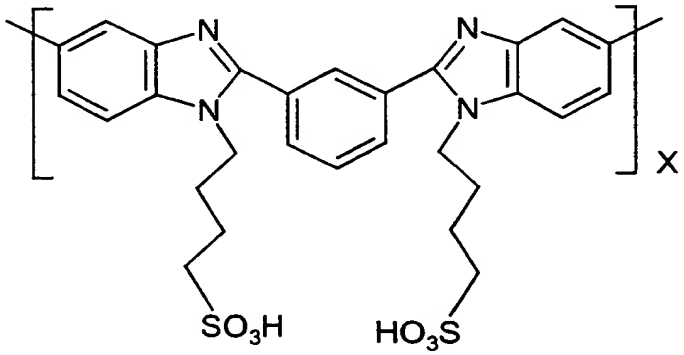
average of the thickness was 47 micrometers.

Example 4

A polybenzimidazole bridged membrane of Example 3 was immersed in 85 percent by weight phosphoric acid at room temperature for 96 hours to yield a polymer electrolyte membrane. This polymer electrolyte membrane was cut out in a circular piece. Next, it was sandwiched by two sheets of carbon electrodes for a fuel cell of the polymer electrolyte type, which were commercially available, and hot-pressed at 140 °C and 50 kgf/cm^2 (1 kgf is equivalent to about 9.8 Newton) to yield a single cell for fuel battery. When hydrogen and air were introduced into this single cell for generating electricity, following outputs were obtained: 300 mW/ cm^2 at 160 °C and 0.5 V under 1 atmospheric pressure, and 475 mW/ cm^2 at 160 °C and 0.5 V under 3 atmospheric pressures, respectively.

Comparative Example 1

Polybenzimidazole having a sulfobutylene group (-$(CH_2)_4$-SO_3H) in a side chain was prepared in a method of Example 1 of U.S. Pat. 5,599,639. Namely, polybenzimidazole having the following repeating unit was prepared.



Poly 2,2'-(m-phenylene)-5,5'-bibenzimidazole was dissolved in N-methyl-2-pyrrolidone, and then lithium hydride was added to the solution. Subsequently, the solution was heated to 70 °C for 12 hours under stirring. After the bubbling ceased, a mixture of 9 parts by mole of 1,4-butanesultone and N-methyl-2-pyrrolidone were added dropwise by an addition funnel. Subsequently, the solution was further heated to 70 °C for 12 hours under stirring. Thus, poly 2,2'-(m-phenylene)-N, N'-sulfobutylene-5,5'-bibenzimidazole having a sulfonation rate of 70.5 percent was obtained.

The polymer was dissolved in dimethylsulfoxide to obtain a polymer solution having a polymer concentration of 5 percent by weight. The solution was cast onto a 6-inch silicon wafer, and dried at 90 °C for four hours in a reduced pressure (10 mmHg) for forming a membrane and for proceeding the bridging reaction at the same time to obtain a cast film. When the cast film thus obtained was immersed in pure water, the cast film considerably swelled so that the film configuration was not kept. The film could not be recovered from the water.

Example 5

To 15 gram of the polymer solution of Comparative Example 1 was added 0.0912 gram of ethyleneglycoldiglycidylether, and mixed thereof. In the solution, a molar ratio of the unsubstituted imidazole group to ethyleneglycoldiglycidylether was about 2:1. The mixed solution thus obtained was cast onto a 6-inch silicon wafer, and dried at 90 °C for four hours in a reduced pressure (10 mmHg) for forming a membrane and for proceeding the bridging reaction at the same time to obtain a cast film. When the cast film thus obtained was immersed in pure water, the film swelled, and it was easily peeled off from the silicon wafer while the film kept the film configuration. A thickness during containing water was about 65 micrometer.

The proton conductivity of the bridged, butylsulfonated polybenzimidazole thus obtained in water was measured by a complex impedance process, and it was 51 mS/cm at 25·C and 118 mS/cm at 57°C.

Therefore, it is confirmed that the present invention may be applied to a basic polymer having proton conductivity.

In the present invention, shaping a basic polymer into a membrane configuration and then proceeding a bridging reaction allow to improve the mechanical strength of the basic polymer. Adjusting an amount of the bridging agent allows to maintain a capacity of the basic polymer to impregnate a strong acid.

Claims:

1. A method for producing a bridged polymer membrane, comprising the steps of:

obtaining a liquid medium comprising a basic polymer having an amino group in a repeating unit, a bridging agent, and a solvent;

shaping the liquid medium into a membrane configuration to obtain a shaped membrane; and

bridging the basic polymer by the bridging agent in the shaped membrane.

2. A method of claim 1 wherein the bridging agent has at least two epoxy groups or isocyanate groups in the molecule thereof.

3. A method of claim 1 or 2 wherein the liquid medium contains 0.001 to 0.8 mole of the bridging agent per unit of the basic polymer.

4. A method of claim 1, 2 or 3 wherein the basic polymer is selected from the group consisting of polybenzimidazoles, polyimidazoles, polyvinylimidazoles, and polybenzbisimidazoles.

5. A method of claim 1 further comprising the step of impregnating the basic polymer with a strong acid for providing proton conductivity.

6. A method of claim 1 wherein the basic polymer has a strong acid group in the repeating unit in the basic polymer.

7. A fuel cell comprising a plurality of single cells, each of the single cells comprising a bridged polymer membrane obtained by a method of claim 5 or 6 and a pair of electrodes sandwiching the bridged polymer membrane.

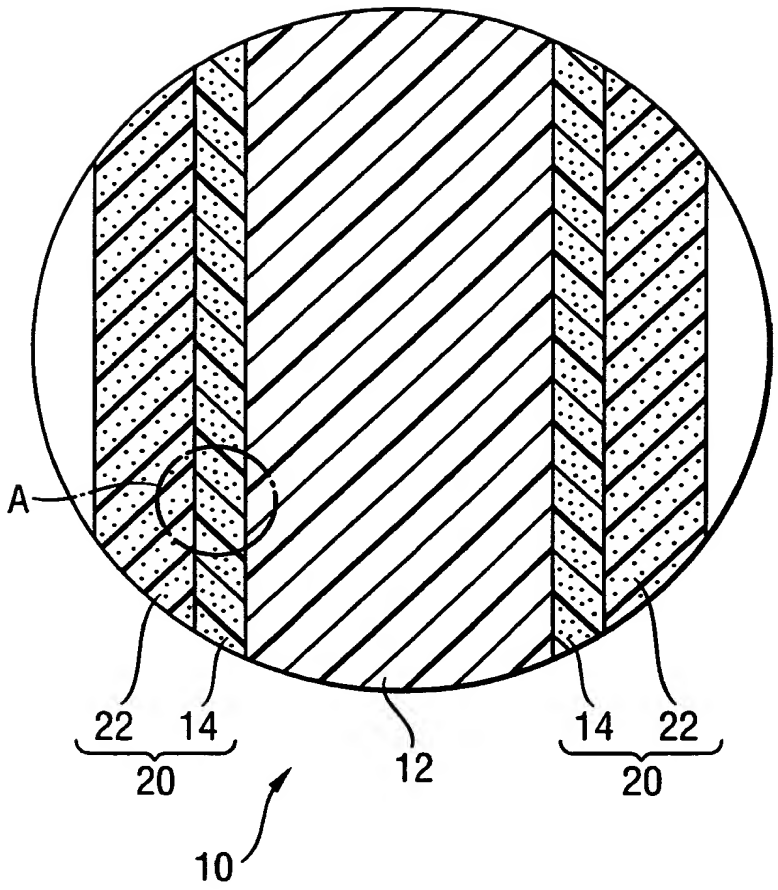


Fig. 1

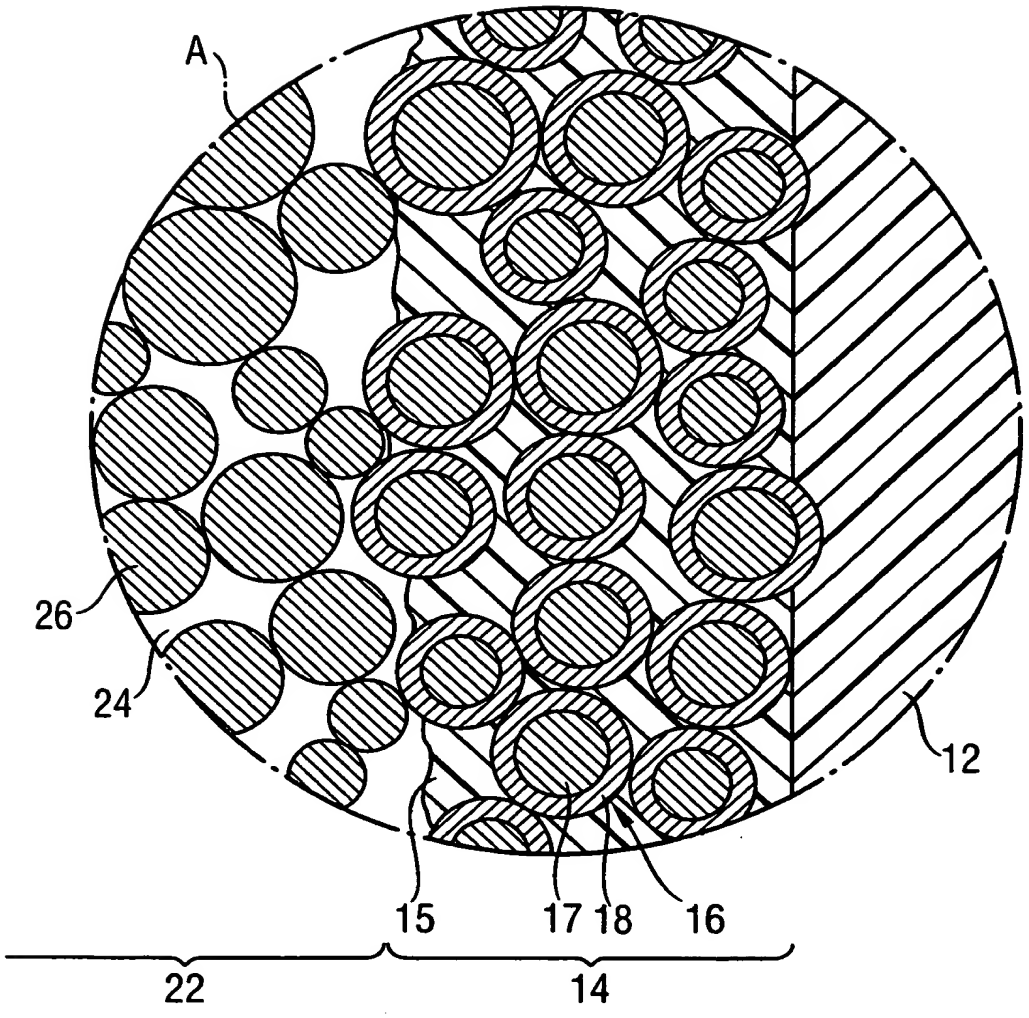


Fig. 2

INTERNATIONAL SEARCH REPORT

In Application No
PCT/[redacted]00/00280

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C08J5/22 H01M8/10

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C08J H01M C25B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 429 759 A (ANDRIEU XAVIER ET AL) 4 July 1995 (1995-07-04) claim 1 column 3, line 25 - line 33 examples	1,2
A	EP 0 025 973 A (ALIGENA AG) 1 April 1981 (1981-04-01) claims 1,5,11	1
A	EP 0 419 396 A (DOW DANMARK) 27 March 1991 (1991-03-27) claims 1,7	1

-/—

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

* Special categories of cited documents :

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
18 May 2000	29/05/2000

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Niaounakis, M

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INTERNATIONAL SEARCH REPORT

In[redacted]pplication No
PCT/EP 00/00280

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
A	DATABASE WPI Section Ch, Week 199734 Derwent Publications Ltd., London, GB; Class A91, AN 1997-369566 XP002138059 & JP 09 157412 A (TOKUYAMA SODA KK), 17 June 1997 (1997-06-17) abstract	1
A	US 5 409 785 A (NAKANO NOBORU ET AL) 25 April 1995 (1995-04-25) claim 1	1

Form PCT/ISA/210 (continuation of second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

Information [redacted] patent family members

Int. [redacted] pplication No
PCT/EP 00/00280

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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PATENT COOPERATION TREATY

PCT

NOTIFICATION OF THE RECORDING OF A CHANGE

(PCT Rule 92bis.1 and Administrative Instructions, Section 422)

From the INTERNATIONAL BUREAU

Eingang 23. April 2001

To:

CELANESE VENTURES GMBH
Intellectual Property Group
Industriepark Höchst, Gebäude K 801
D-65926 Frankfurt am Main
ALLEMAGNE

Date of mailing (day/month/year) 20 March 2001 (20.03.01)	
Applicant's or agent's file reference 1999/FO12 PCT	**IMPORTANT NOTIFICATION**
International application No. PCT/EP00/00280	**International filing date** (day/month/year) 15 January 2000 (15.01.00)

1. The following indications appeared on record concerning:

[X] the applicant [ ] the inventor [ ] the agent [ ] the common representative

Name and Address	State of Nationality	State of Residence
AXIVA GMBH D-65926 Frankfurt am Main Germany	DE	DE
	Telephone No. 069-305-4302	
	Facsimile No. 069-305-26600	
	Teleprinter No.	

2. The International Bureau hereby notifies the applicant that the following change has been recorded concerning:

[ ] the person [X] the name [ ] the address [ ] the nationality [ ] the residence

Name and Address	State of Nationality	State of Residence
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3. Further observations, if necessary:
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4. A copy of this notification has been sent to:

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1999/F-012

ANTRAG (4) pgs.

Der Unterzeichnete beantragt, daß die vorliegende internationale Anmeldung nach dem Vertrag über die internationale Zusammenarbeit auf dem Gebiet des Patentwesens behandelt wird.

V m Anmeldeamt auszufüllen

Internationales Aktenzeichen: PCT/EP 0 0 / 0 0 2 8 0

Internationales Anmeldedatum: 15 JAN 2000 (15.01.00)

EUROPEAN PATENT OFFICE
PCT INTERNATIONAL APPLICATION
Name des Anmeldeamts und "PCT International Application"

Aktenzeichen des Anmelders oder Anwalts *(falls gewünscht)* *(max. 12 Zeichen)*: 1999/F012 PCT

Feld Nr. I BEZEICHNUNG DER ERFINDUNG

Method for producing bridged polymer membrane and fuel cell

Feld Nr. II ANMELDER

Name und Anschrift: *(Familienname, Vorname; bei juristischen Personen vollständige amtliche Bezeichnung. Bei der Anschrift sind die Postleitzahl und der Name des Staats anzugeben. Der in diesem Feld in der Anschrift angegebene Staat ist der Staat des Sitzes oder Wohnsitzes des Anmelders, sofern nachstehend kein Staat des Sitzes oder Wohnsitzes angegeben ist.)*

Axiva GmbH

D-65926 Frankfurt am Main
Deutschland

☐ Diese Person ist gleichzeitig Erfinder

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Telefaxnr.: 069-305-26600

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Staatsangehörigkeit (Staat): DE

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Japan

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- ☐ LV Lettland
- ☐ MD Republik Moldau
- ☐ MG Madagaskar
- ☐ MK Die ehemalige jugoslawische Republik Mazedonien
- ☐ MN Mongolei
- ☐ MW Malawi
- ☒ MX Mexiko
- ☐ NO Norwegen
- ☐ NZ Neuseeland
- ☐ PL Polen
- ☐ PT Portugal
- ☐ RO Rumänien
- ☐ RU Russische Föderation
- ☐ SD Sudan
- ☐ SE Schweden
- ☐ SG Singapur
- ☐ SI Slowenien
- ☐ SK Slowakei
- ☐ SL Sierra Leone
- ☐ TJ Tadschikistan
- ☐ TM Turkmenistan
- ☐ TR Türkei
- ☐ TT Trinidad und Tobago
- ☐ UA Ukraine
- ☐ UG Uganda
- ☒ US Vereinigte Staaten von Amerika
- ☐ UZ Usbekistan
- ☐ VN Vietnam
- ☐ YU Jugoslawien
- ☐ ZW Simbabwe

Kästchen für die Bestimmung von Staaten (für die Zwecke eines nationalen Patents), die dem PCT nach der Veröffentlichung dieses Formblatts beigetreten sind:

- ☐ AE Vereinigte arabische Emirate
- ☒ ZA Südafrika
- ☐

Erklärung bzgl. vorsorglicher Bestimmungen: Zusätzlich zu den oben genannten Bestimmungen nimmt der Anmelder nach Regel 4.9 Absatz b auch alle anderen nach dem PCT zulässigen Bestimmungen vor mit Ausnahme der im Zusatzfeld genannten Bestimmungen, die v n dieser Erklärung ausgenommen sind. Der Anmelder erklärt, daß diese zusätzlichen Bestimmungen unter dem Vorbehalt einer Bestätigung stehen und jede zusätzliche Bestimmung, die vor Ablauf von 15 Monaten ab dem Prioritätsdatum nicht bestätigt wurde, nach Ablauf dieser Frist als vom Anmelder zurückgenommen gilt. *(Die Bestätigung einer Bestimmung erfolgt durch die Einreichung einer Mitteilung, in der diese Bestimmung angegeben wird, und die Zahlung der Bestimmungs- und der Bestätigungsgebühr. Die Bestätigung muß beim Anmeldeamt innerhalb der Frist von 15 Monaten eingehen.)*

Feld Nr. VI PRIORITÄTSANSPRUCH ☐ Weitere Prioritätsansprüche sind im Zusatzfeld angegeben.

Anmeldedatum der früheren Anmeldung *(Tag/Monat)*	Aktenzeichen der früheren Anmeldung	Ist die frühere Anmeldung eine: national Anmeldung: Staat	regionale Anmeldung:* regionales Amt	internati nale Anmeldung: Anmeldeamt
Zeile (1) 27. Januar 1999 (27.01.99)	11-018090	JP		
Zeile (2) 04. Juni 1999 (04.06.99)	11-157509	JP		
Zeile (3)				

☐ Das Anmeldeamt wird ersucht, eine beglaubigte Abschrift der oben in der (den) Zeile(n) _____ bezeichneten früheren Anmeldung(en) zu erstellen und dem internationalen Büro zu übermitteln *(nur falls die frühere Anmeldung(en) bei dem Amt eingereicht worden ist(sind), das für die Zwecke dieser internationalen Anmeldung Anmeldeamt ist)*

** Falls es sich bei der früheren Anmeldung um eine ARIPO-Anmeldung handelt, so muß in dem Zusatzfeld mindestens ein Staat angegeben werden, der Mitgliedstaat der Pariser Verbandsübereinkunft zum Schutz des gewerblichen Eigentums ist und für den die frühere Anmeldung eingereicht wurde.*

Feld Nr. VII INTERNATIONALE RECHERCHENBEHÖRDE

Wahl der internationalen Recherchenbehörde (ISA) *(falls zwei oder mehr als zwei internationale Recherchenbehörden für die Ausführung der internationalen Recherche zuständig sind, geben Sie die von Ihnen gewählte Behörde an; der Zweibuchstaben-Code kann benutzt werden)*

ISA /

Antrag auf Nutzung der Ergebnisse einer früheren Recherche; Bezugnahme auf diese frühere Recherche *(falls eine frühere Recherche bei der internationalen Recherchenbehörde beantragt oder von ihr durchgeführt worden ist):*

Datum *(Tag/Monat/Jahr)* Aktenzeichen Staat *(oder regionales Amt)*

Feld Nr. VIII KONTROLLISTE; EINREICHUNGSSPRACHE

Diese internationale Anmeldung enthält die folgende **Anzahl von Blättern:**

Antrag	: 3
Beschreibung (ohne Sequenzprotokollteil)	: 18
Ansprüche	: 1
Zusammenfassung	: 1
Zeichnungen	: 2
Sequenzprotokollteil der Beschreibung	:
Blattzahl insgesamt	: 25

Dieser internationalen Anmeldung liegen die nachstehend angekreuzten Unterlagen bei:

1. ☒ Blatt für die Gebührenberechnung
2. ☐ Gesonderte unterzeichnete Vollmacht
3. ☐ Kopie der allgemeinen Vollmacht; Aktenzeichen (falls vorhanden):
4. ☐ Begründung für das Fehlen einer Unterschrift
5. ☒ Prioritätsbeleg(e), in Feld Nr. VI durch folgende Zeilennummer gekennzeichnet:
6. ☐ Übersetzung der internationalen Anmeldung in die folgende Sprache:
7. ☐ Gesonderte Angaben zu hinterlegten Mikroorganismen oder anderem biologischen Material
8. ☐ Sequenzprotokolle für Nucleotide und/oder Aminosäuren in computerlesbarer Form
9. ☒ Sonstige *(einzeln aufführen)*: Übersetzung der Pris. Belege

Abbildung der Zeichnungen, die mit der Zusammenfassung veröffentlicht werden soll (Nr.):

Sprache, in der die internationale Anmeldung eingereicht wird: englisch

Feld Nr. IX UNTERSCHRIFT DES ANMELDERS ODER DES ANWALTS

Der Name jeder unterzeichnenden Person ist neben der Unterschrift zu wiederholen, und es ist anzugeben, sofern sich dies nicht eindeutig aus dem Antrag ergibt, in welcher Eigenschaft die Person unterzeichnet.

Axiva GmbH

ppa. Auch i.V. Dr. Popp

Vom Anmeldeamt auszufüllen

1. Datum des tatsächlichen Eingangs dieser internationalen Anmeldung: 15 JAN 2000 (15. 01. 00)

2. Zeichnungen ☒ eingegangen: ☐ nicht eingegangen:

3. Geändertes Eingangsdatum aufgrund nachträglich, jedoch fristgerecht eingegangener Unterlagen oder Zeichnungen zur Vervollständigung dieser internationalen Anmeldung:

4. Datum des fristgerechten Eingangs der angeforderten Richtigstellungen nach Artikel 11(2) PCT:

5. Internati nale Recherchenbehörde *(falls zwei oder mehr zuständig sind)*: ISA /

6. ☐ Übermittlung des Recherchenexemplars bis zur Zahlung der Recherchengebühr aufgeschoben

Vom Internationalen Büro auszufüllen

Datum des Eingangs des Aktenexemplars beim Internationalen Büro:

Zusatzfeld *Wird dieses Zusatzfeld nicht benutzt, so sollte dieses Blatt dem Antrag nicht beigefügt werden.*

1. Wenn der ***Platz in einem Feld nicht für alle Angaben ausreicht****: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. ..." [Nummer des Feldes angeben] und machen die Angaben entsprechend der in dem Feld, in dem der Platz nicht ausreicht, vorgeschriebenen Art und Weise, insbesondere:*

(i) ***Wenn mehr als zwei Anmelder und/oder Erfinder vorhanden sind*** *und kein "Fortsetzungsblatt" zur Verfügung steht: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. III" und machen für jede weitere Person die in Feld Nr. III vorgeschriebenen Angaben. Der in diesem Feld in der Anschrift angegebene Staat ist der Staat des Sitzes oder Wohnsitzes des Anmelders, sofern nachstehend kein Staat des Sitzes oder Wohnsitzes angegeben ist.*

(ii) Wenn in Feld Nr. II oder III die Angabe ***"die im Zusatzfeld angegebenen Staaten"*** *angekreuzt ist: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. II", "Fortsetzung von Feld Nr. III" bzw. "Fortsetzung von Feld Nr. II und Nr. III" und geben den Namen des Anmelders oder die Namen der Anmelder an und neben jedem Namen den Staat oder die Staaten (und/oder ggf. ARIPO-, eurasisches, europäisches oder OAPI-Patent), für die die bezeichnete Person Anmelder ist.*

(iii) Wenn der in Feld Nr. II oder III genannte ***Erfinder oder Erfinder/Anmelder nicht für alle Bestimmungsstaaten oder für die Vereinigten Staaten von Amerika als Erfinder benannt ist****: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. II", "Fortsetzung von Feld Nr. III" bzw. "Fortsetzung von Feld Nr. II und Nr. III" und geben den Namen des Erfinders oder die Namen der Erfinder an und neben jedem Namen den Staat oder die Staaten (und/oder ggf. ARIPO-, eurasisches, europäisches oder OAPI-Patent), für die die bezeichnete Person Erfinder ist.*

(iv) Wenn zusätzlich zu dem Anwalt oder den Anwälten, die in Feld Nr. IV angegeben sind, ***weitere Anwälte*** *bestellt sind: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. IV" und machen für jeden weiteren Anwalt die entsprechenden, in Feld Nr. IV vorgeschriebenen Angaben.*

(v) Wenn in Feld Nr. V bei einem Staat (oder bei OAPI) die Angabe ***"Zusatzpatent"*** *oder* ***"Zusatzzertifikat,"*** *oder wenn in Feld Nr. V bei den Vereinigten Staaten von Amerika die Angabe* ***"Fortsetzung"*** *oder* ***"Teilfortsetzung"*** *hinzugefügt wird: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. V" und geben den Namen des betreffenden Staats (oder OAPI) an und nach dem Namen jedes solchen Staats (oder OAPI) das Aktenzeichen des Hauptschutzrechts oder der Hauptschutzrechtsanmeldung und das Datum der Erteilung des Hauptschutzrechts oder der Einreichung der Hauptschutzrechtsanmeldung.*

(vi) Wenn in Feld Nr. VI ***die Priorität von mehr als drei früheren Anmeldungen beansprucht wird****: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. VI" und machen für jede weitere frühere Anmeldung die entsprechenden, in Feld Nr. VI vorgeschriebenen Angaben.*

(vii) Wenn in Feld Nr. VI die ***frühere Anmeldung eine ARIPO Anmeldung*** *ist: In diesem Fall schreiben Sie "Fortsetzung von Feld Nr. VI" und geben, unter Angabe der Nummer der Zeile, in der die die frühere Anmeldung betreffenden Angaben gemacht sind, mindestens einen Staat an, der Mitglied der Pariser Verbandsübereinkunft zum Schutz des gewerblichen Eigentums ist und für den die frühere Anmeldung erfolgte.*

2. Wenn, im Hinblick auf die ***Erklärung bzgl. vorsorglicher Bestimmungen*** *in Feld Nr. V, der Anmelder Staaten von dieser Erklärung ausnehmen möchte: In diesem Fall schreiben Sie "Bestimmung(en), die von der Erklärung bzgl. vorsorglicher Bestimmungen ausgenommen ist(sind)" und geben den Namen oder den Zweibuchstaben-Code jedes so ausgeschlossenen Staates an.*

3. Wenn der Anmelder für irgendein Bestimmungsamt die Vorteile nationaler Vorschriften betreffend ***unschädliche Offenbarung oder Ausnahmen von der Neuheitsschädlichkeit*** *in Anspruch nimmt: In diesem Fall schreiben Sie "Erklärung betreffend unschädliche Offenbarung oder Ausnahmen von der Neuheitsschädlichkeit" und geben im folgenden die entsprechende Erklärung ab.*

Feld Nr. IX. Unterschrift des Anmelder oder des Anwalts

Tetsu Yamamoto

Tetsu Yamamoto

PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference	FOR FURTHER ACTION	see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.
1999/F012 PCT		
International application No.	International filing date *(day/month/year)*	(Earliest) Priority Date *(day/month/year)*
PCT/EP 00/ 00280	15/01/2000	27/01/1999
Applicant		
AXIVA GMBH et al.		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 3 sheets.

[X] It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**

   a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

   [ ] the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

   b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing :

   [ ] contained in the international application in written form.

   [ ] filed together with the international application in computer readable form.

   [ ] furnished subsequently to this Authority in written form.

   [ ] furnished subsequently to this Authority in computer readble form.

   [ ] the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

   [ ] the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. [ ] **Certain claims were found unsearchable** (See Box I).

3. [ ] **Unity of invention is lacking** (see Box II).

4. With regard to the **title,**

   [X] the text is approved as submitted by the applicant.

   [ ] the text has been established by this Authority to read as follows:

5. With regard to the **abstract,**

   [X] the text is approved as submitted by the applicant.

   [ ] the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No. _____

   [ ] as suggested by th applicant.

   [ ] because the applicant failed to suggest a figure.

   [ ] because this figure better characterizes the invention.

   [X] None of the figures.

INTERNATIONAL SEARCH REPORT

International Application No
P[redacted]P 00/00280

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C08J5/22 H01M8/10

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C08J H01M C25B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 429 759 A (ANDRIEU XAVIER ET AL) 4 July 1995 (1995-07-04) claim 1 column 3, line 25 - line 33 examples	1,2
A	EP 0 025 973 A (ALIGENA AG) 1 April 1981 (1981-04-01) claims 1,5,11	1
A	EP 0 419 396 A (DOW DANMARK) 27 March 1991 (1991-03-27) claims 1,7	1

-/--

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
18 May 2000	29/05/2000

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31–70) 340–2040, Tx. 31 651 epo nl, Fax: (+31–70) 340–3016	Niaounakis, M

1

Form PCT/ISA/210 (second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

International Application No
P[redacted]P 00/00280

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
A	DATABASE WPI Section Ch, Week 199734 Derwent Publications Ltd., London, GB; Class A91, AN 1997-369566 XP002138059 & JP 09 157412 A (TOKUYAMA SODA KK), 17 June 1997 (1997-06-17) abstract	1
A	US 5 409 785 A (NAKANO NOBORU ET AL) 25 April 1995 (1995-04-25) claim 1	1

1

Form PCT/ISA/210 (continuation of second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
PCT/EP 00/00280

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 5429759 A	04-07-1995	FR 2695131 A	04-03-1994
		AT 156937 T	15-08-1997
		CA 2105231 A	02-03-1994
		DE 69313063 D	18-09-1997
		DE 69313063 T	11-12-1997
		EP 0591014 A	06-04-1994
		ES 2105164 T	16-10-1997
		JP 6196016 A	15-07-1994
EP 0025973 A	01-04-1981	BR 8005980 A	31-03-1981
		CA 1186860 A	14-05-1985
		DE 3035134 A	09-04-1981
		DE 3070021 D	07-03-1985
		FR 2465508 A	27-03-1981
		GB 2058798 A,B	15-04-1981
		IL 61070 A	31-05-1984
		JP 1600018 C	31-01-1991
		JP 2023215 B	23-05-1990
		JP 56048214 A	01-05-1981
		MX 156797 A	05-10-1988
		US 4584103 A	22-04-1986
EP 0419396 A	27-03-1991	BR 9004424 A	10-09-1991
		CA 2024575 A	06-03-1991
		JP 3114518 A	15-05-1991
		NZ 235165 A	26-03-1993
JP 9157412 A	17-06-1997	NONE	
US 5409785 A	25-04-1995	JP 5174856 A	13-07-1993
		CA 2085549 A	26-06-1993
		DE 4243600 A	01-07-1993
		JP 6076838 A	18-03-1994

PATENT COOPERATION TREATY

From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

AXIVA GMBH
Patente
Industriepark Höchst, Gebäude K 801
D-65926 Frankfurt am Main
ALLEMAGNE

PCT

NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL PRELIMINARY EXAMINATION REPORT
(PCT Rule 71.1)

Date of mailing *(day/month/year)*	26.02.2001
Applicant's or agent's file reference 1999/FO12	**IMPORTANT NOTIFICATION**

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/EP00/00280	15/01/2000	27/01/1999

Applicant
AXIVA GMBH et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.

2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.

3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

4. **REMINDER**

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Name and mailing address of the IPEA/	Authorized officer
European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Aperribay, I Tel.+49 89 2399-8154

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
1999/FO12		
International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/EP00/00280	15/01/2000	27/01/1999
International Patent Classification (IPC) or national classification and IPC C08J5/22		
Applicant AXIVA GMBH et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 5 sheets, including this cover sheet.

   ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

   - I ☒ Basis of the report
   - II ☐ Priority
   - III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - IV ☐ Lack of unity of invention
   - V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations suporting such statement
   - VI ☐ Certain documents cited
   - VII ☒ Certain defects in the international application
   - VIII ☒ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
17/08/2000	26.02.2001
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Kairi, M Telephone No. +49 89 2399 8672

Form PCT/IPEA/409 (cover sheet) (January 1994)

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/EP00/00280

I. Basis of the r port

1. This report has been drawn on the basis of *(substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments (Rules 70.16 and 70.17).)*:
Description, pages:

1-16 as originally filed

Claims, No.:

1-7 as originally filed

Drawings, sheets:

1/2-2/2 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:

☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	3,4,7
	No:	Claims	1,2,5,6
Inventive step (IS)	Yes:	Claims	4
	No:	Claims	3,7
Industrial applicability (IA)	Yes:	Claims	1-7
	No:	Claims	

2. Citations and explanations
see separate sheet

VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:
se separate sheet

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:
see separate sheet

R lt m V

Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

Article 33(2) PCT

The document US-A-5,429,759 (D1) discloses a proton-conducting polymer solid electrolyte comprising a polyether which has two terminal primary amine functions and which is cross-linked by a curing agent having two epoxy functions, said electrolyte being characterized by the fact that an acid is dissolved into said cross-linked polyether and that said electrolyte contains water (Claim 1). That electrolyte is particularly suitable for use in numerous applications, such as electrochemical cells, fuel cells, electrolytic cells, supercapacitors, electrochemical sensors, electrochromic windows and displays, and in all microionic applications that use solid electrolytes (Column 3, lines 25-33). Example 1 discloses the following:
The following were dissolved in acetonitrile at 24°C:
4 grams of O,O'-bis-(2-aminopropyl)-polyethylene glycol 6000 (Jeffamine ED 6000 sold by Texaco);
0.08 g of butanediol diglycidyl ether as a curing agent; and
0.08 g of LiClO4 as a catalyst.
After complete dissolution, the mixture was cast into a mold, and then the solvent was evaporated in vacuo. The resulting film was cross-linked for 15 hours at 100°C.
The cross-linked film was then swollen with an aqueous phosphoric solution, and it was then dried in vacuo to remove the water the water for 15 hours at 100°C.
The resulting anhydrous polymer solid electrolyte contained 30% by weight of phosphoric acid.
Example 4 discloses the following:
By following the same procedure, a cross-linked polymer film was made from:
2 g of O,O'-bis-(2-aminopropyl)-polyethylene glycol 1900 (Jeffamine ED 2001):
0.4 g of butanediol diglycidyl ether; and
0.2 g of paratoluene-sulfonic acid.
The cross-linked film was then dried for 15 hours at 100°C under a primary vacuum.
On the basis of that disclosure the subject-matter of Claims 1,2, 5 and 6 is anticipated by D1.

Article 33(3) PCT

Regarding the subject-matter of Claim 3, the individual feature of that claim could correspond to an arbitrary selection in the absence of evidence of unexpected effect and therefore no inventive step can be recognized for the subject-matter of that claim.

Regarding the subject-matter of Claim 4, the skilled man starting from the disclosure in D1 would not have obviously considered using as a basic polymer having an amino group a polymer having an imidazole compound repeating unit with a secondary amino group and therefore an inventive step can be recognized for the subject-matter of that claim.

Regarding the subject-matter of Claim 7, the individual feature of that claim can be considered to be conventional as also indicated from the decription (page 1, lines 14-15) and therefore no inventive step can be recognized for the subject-matter of that claim.

Re Item VII

Certain defects in the international application

The statement of prior art does not include a reference to the closest prior art document US-A-5 429 759 (D1) (Rule 5.1(a)(ii) PCT).

Re Item VIII

Certain observations on the international application

The "incorporation as reference" of the entire content of another document (page 2, lines 9-10 and 24-25; page 7, lines 4-5; page 10, lines 12-13) could cast doubt upon the protection given by the claims (Article 6 PCT).

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PATENT COOPERATION TREATY

From the INTERNATIONAL BUREAU

PCT

NOTICE INFORMING THE APPLICANT OF THE COMMUNICATION OF THE INTERNATIONAL APPLICATION TO THE DESIGNATED OFFICES

(PCT Rule 47.1(c), first sentence)

To:
AXIVA GMBH
Patente
Industriepark Höchst, Gebäude K 801
D-65926 Frankfurt am Main
ALLEMAGNE

Date of mailing (day/month/year) 03 August 2000 (03.08.00)	
Applicant's or agent's file reference 1999/FO12 PCT	**IMPORTANT NOTICE**

International application No.	**International filing date** (day/month/year)	**Priority date** (day/month/year)
PCT/EP00/00280	15 January 2000 (15.01.00)	27 January 1999 (27.01.99)

Applicant
AXIVA GMBH et al

1. Notice is hereby given that the International Bureau has communicated, as provided in Article 20, the international application to the following designated Offices on the date indicated above as the date of mailing of this Notice:
   JP,KR,US

- In accordance with Rule 47.1(c), third sentence, those Offices will accept the present Notice as conclusive evidence that the communication of the international application has duly taken place on the date of mailing indicated above and no copy of the international application is required to be furnished by the applicant to the designated Office(s).

2. The following designated Offices have waived the requirement for such a communication at this time:
   CA,EP,MX,ZA

   The communication will be made to those Offices only upon their request. Furthermore, those Offices do not require the applicant to furnish a copy of the international application (Rule 49.1(a-bis)).

3. Enclosed with this Notice is a copy of the international application as published by the International Bureau on
   03 August 2000 (03.08.00) under No. WO 00/44816

REMINDER REGARDING CHAPTER II (Article 31(2)(a) and Rule 54.2)

If the applicant wishes to postpone entry into the national phase until 30 months (or later in some Offices) from the priority date, **a demand for international preliminary examination** must be filed with the competent International Preliminary Examining Authority before the expiration of 19 months from the priority date.

It is the applicant's sole responsibility to monitor the 19-month time limit.

Note that only an applicant who is a national or resident of a PCT Contracting State which is bound by Chapter II has the right to file a demand for international preliminary examination.

REMINDER REGARDING ENTRY INTO THE NATIONAL PHASE (Article 22 or 39(1))

If the applicant wishes to proceed with the international application in the **national phase**, he must, within 20 months or 30 months, or later in some Offices, perform the acts referred to therein before each designated or elected Office.

For further important information on the time limits and acts to be performed for entering the national phase, see the Annex to Form PCT/IB/301 (Notification of Receipt of Record Copy) and Volume II of the PCT Applicant's Guide.

The International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland

Facsimile No. (41-22) 740.14.35

Authorized officer
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